Super- and Hyperdeformed Isomeric States and Long-lived Superheavy Elements

- 1

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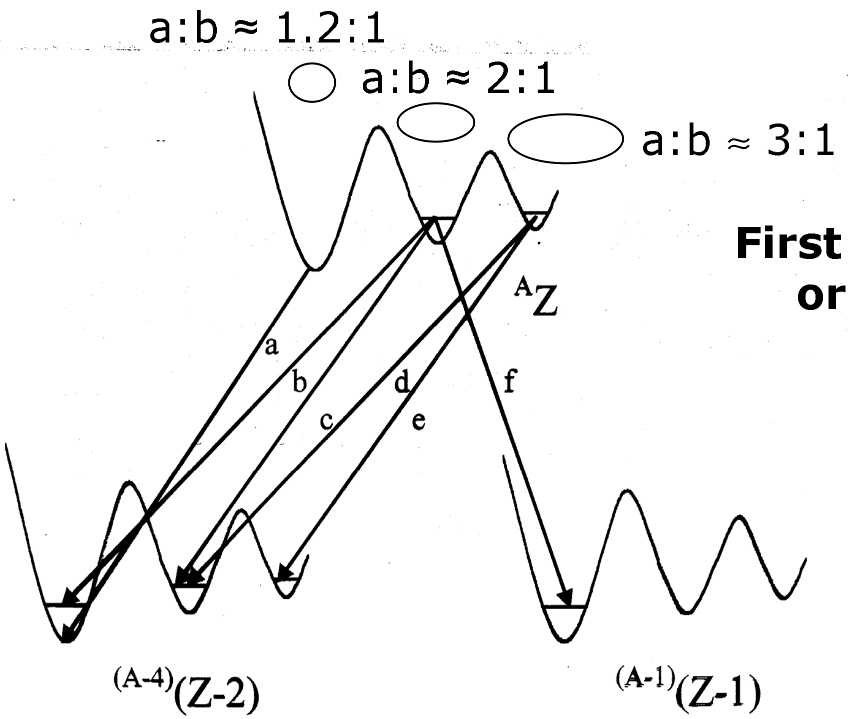
50th Anniversary of the Karlsruhe Nuclide Chart

Collaborators since 1970:

C.J. Batty	Rutherford Laboratory
A.I. Kilvington	Rutherford Laboratory
G.W.A. Newton	Manchester University
V.J. Robinson	Manchester University
J.D. Hemingway	Research Reactor, Risley
J.L. Weil	Kentucky University
A.M. Friedman	Argonne National Laboratory
D.S. Mather	AWRE, Aldermaston, Berkshire
S. Eshhar	The Hebrew University
D. Kolb	Kassel University
S. Gelberg	The Hebrew University
A. Pape	Strasbourg Nuclear Institute
R. Brandt	Marburg University
R.V. Gentry	Knoxville, Tennessee
H. W. Miller	Boulder, Colorado
L. Halicz	Geological Survey of Israel
I. Segal	Geological Survey of Israel
I. Rodushkin	Anlytica, Loleà, Sweden
Y. Kashiv	The Hebrew University

Special thanks I owe to Nissan Zeldes



First minimum: Spherical or normal deformed

Second minimum: Suerdeformed

Third minimum: Hyperdeformed

a: $I^{min} \rightarrow I^{min}$. Normal α's.

b: $II^{min} \rightarrow I^{min}$. Retarded α's: $^{190}Ir \rightarrow {}^{186}Re$.

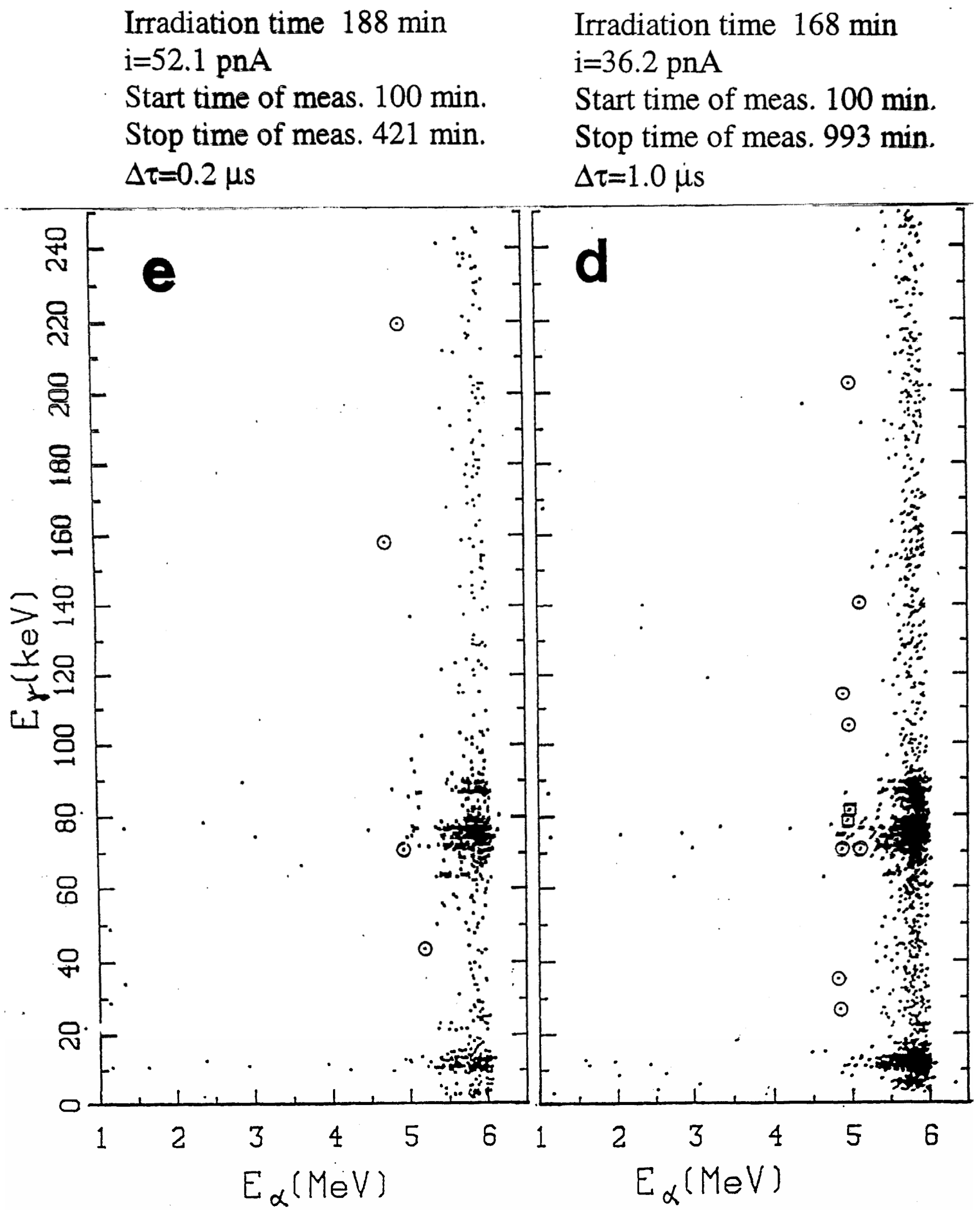
c: $II^{min} \rightarrow II^{min}$. Enhanced α's: $^{210}Fr \rightarrow {}^{106}At$; ~ $^{238}Am \rightarrow {}^{234}Np$.

d: $III^{min} \rightarrow II^{min}$. Retarded α's: $^{195}Hg \rightarrow {}^{191}Pt$.

e: $III^{min} \rightarrow III^{min}$. Enhanced α's: ~ $^{247}Es \rightarrow {}^{243}Bk$; ~ $^{252}No \rightarrow {}^{248}Fm$.

f: $II^{min} \rightarrow I^{min}$. Retarded protons: $^{198}Tl \rightarrow {}^{197}Hg$ (?); $^{205}Fr \rightarrow {}^{204}Rn$ (?).

Fig. 12. Summary of the new kinds of seen transitions and their properties.



$^{16}O + ^{197}Au$ E_{Lab} = 80 MeV

We used catcher foil technique and measured off-line α-γ coincidences from the catcher foil

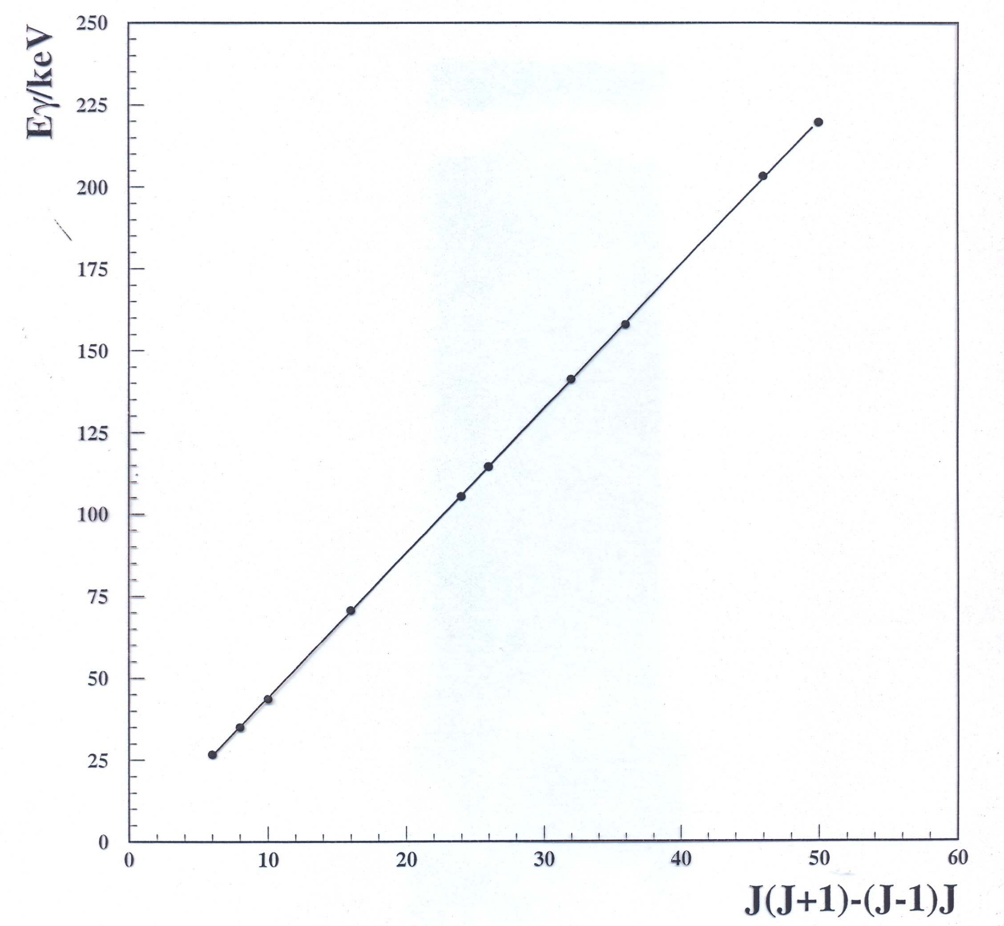
We found 5.2 MeV α-particle group in coincidence with various γ-rays. ($\sigma \approx$ 30 nb)

It was identified as a transition from
^{210}Fr to ^{206}At

Eα for g.s. to g.s. transition is 6.54 MeV; $t_{1/2}$ =190 s.

$t_{1/2}$(Cal.) for 5.2 MeV is 1.6X10^9 s.
Why it decays with low energy when much higher energy with seven orders of magnitude larger transition probability is available?

$t_{1/2}$ (exp.) $\approx$ 90 m
$t_{1/2}$ (Cal.) for 5.2 MeV α-particle from ^{210}Fr = 51 y
It is enhanced by a factor of 3x10^5



E_x = 4.40xJ(J+1)

4.40 keV is characteristic for SDB transitions

Conclusion:

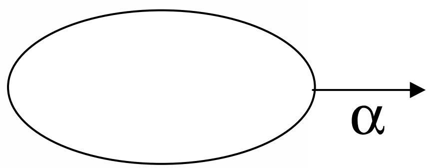
The 5.2 MeV α-particles decay to a

SDB state.

The potential parameters of Igo were used, but

$$R=R_0(1+\beta_2 Y_{20}(\theta)+\beta_3 Y_{30}(\theta)+\beta_4 Y_{40}(\theta))$$

E_α	β_2	β_3	β_4	$t_{1/2}(T)(m)$	$t_{1/2}(T)/t_{1/2}(E)$
6.57	0.0	0.0	0.0	6.0	1.1
5.2	0.0	0.0	0.0	$2.8\text{x}10^7$	$3.1\text{x}10^5$
5.2	0.7	0.0	0.0	$1.3\text{x}10^3$	$1.4\text{x}10^1$
5.2	0.7	0.07	0.0	$3.5\text{x}10^2$	3.9
5.2	0.7	0.15	0.0	$8.2\text{x}10^1$	0.9
5.2	0.7	0.07	0.06	$1.0\text{x}10^2$	1.1



The data can consistently be interpreted in terms of a transition from a high spin state in the SD minimum of the parent nucleus to a high spin state in the SD minimum of the daughter.

Mod. Phys. Letters A11, 861(1996).

Theoretical Predictions SDBH (^{206}At)

W. Satula et al. (1991)
(Macroscopic-Microscopic)
(Strutinsky)

S.J. Krieger et al. (1992)
(Hartree-Fock + BCS)

7.25 MeV (ext.) 10.89 MeV(int.)

α decay to spin 18 at 1.50 (MeV); [Ex=4.40xJ(J+1)]

Ex in ^{206}At

≥8.75 MeV ≥12.39 MeV

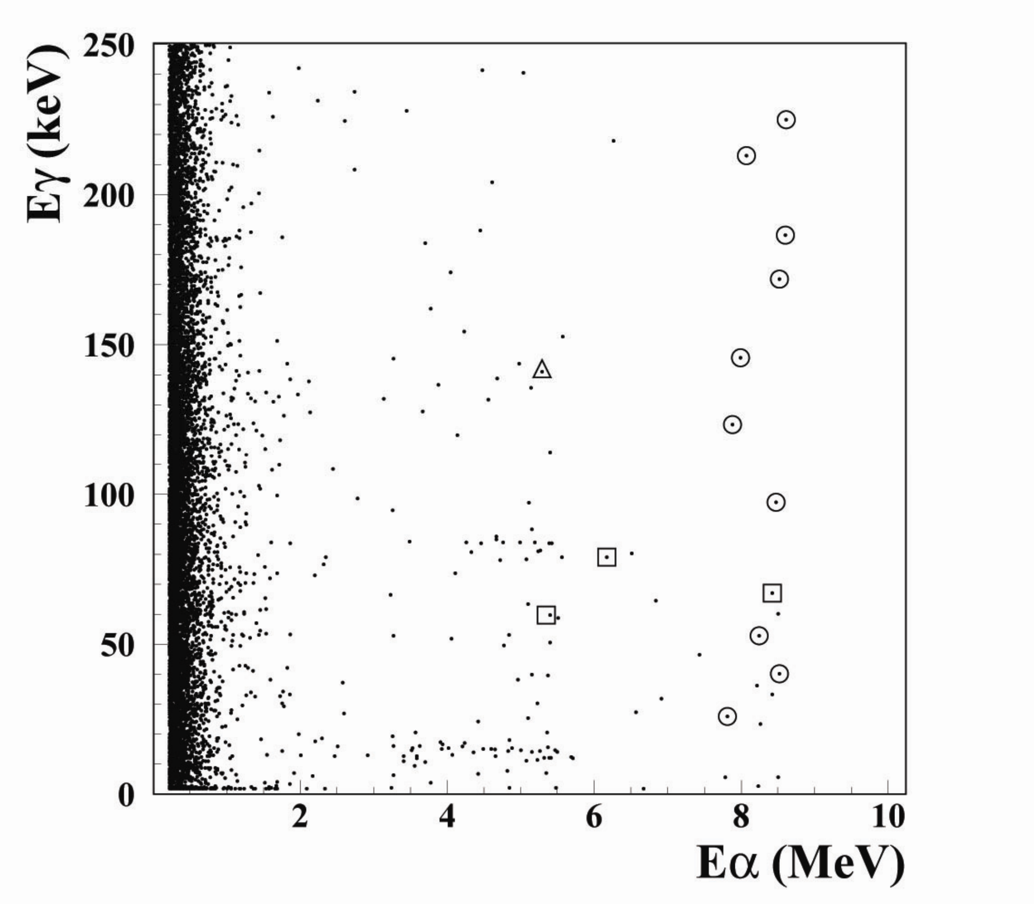
Ex in 210Fr (E_x (^{206}At)+(5.3-6.70))

≥7.35 **≥10.99**

The second experiment we performed using the pelleton accelerator at the Weizmann Institute was:

$^{28}Si + ^{181}Ta$ at $E_{Lab} = 125$ MeV (This is about 10% below the Coulomb barrier)

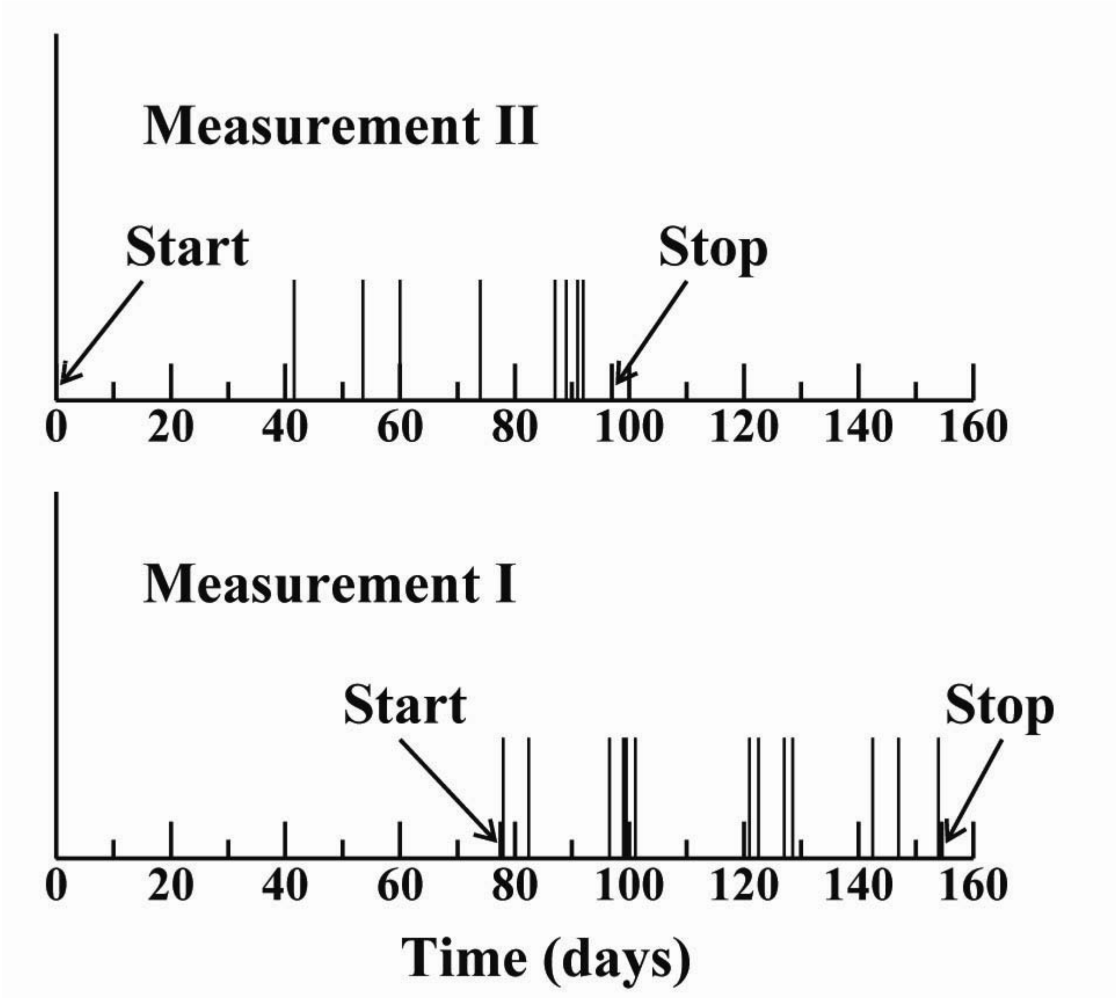
The compound nucleus is ^{209}Fr



Irradiation: T=42.5 h; i=11.5pnA; dose: 1.1×10^{16} particles.

Measurement: T_1=77.4 d; T_2=154.2 d; ΔT=76.8 d.

Catcher foil: 200 μg/cm² of C.

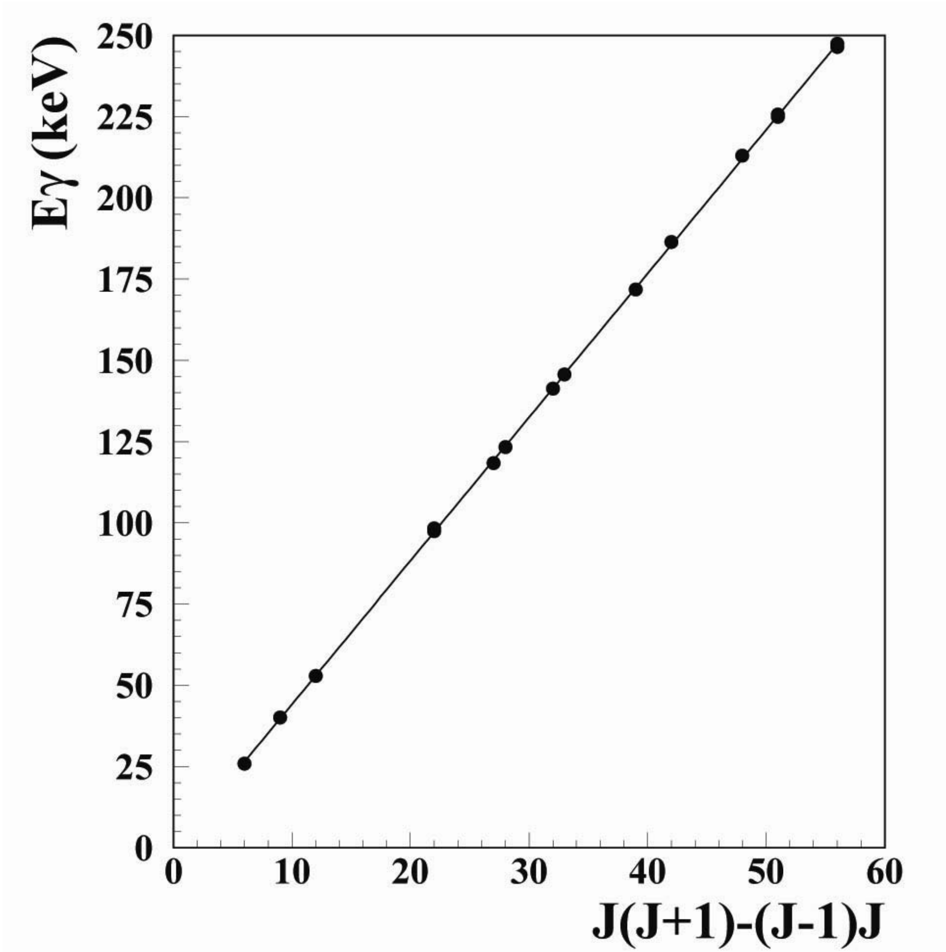


$40\ d \leq t_{1/2} \leq 2\ y$

8.6 MeV is a very high energy for α-particles.

$t_{1/2}$(Cal.) ≈ 1 μs

Retardation factor ≈ 10^{12}



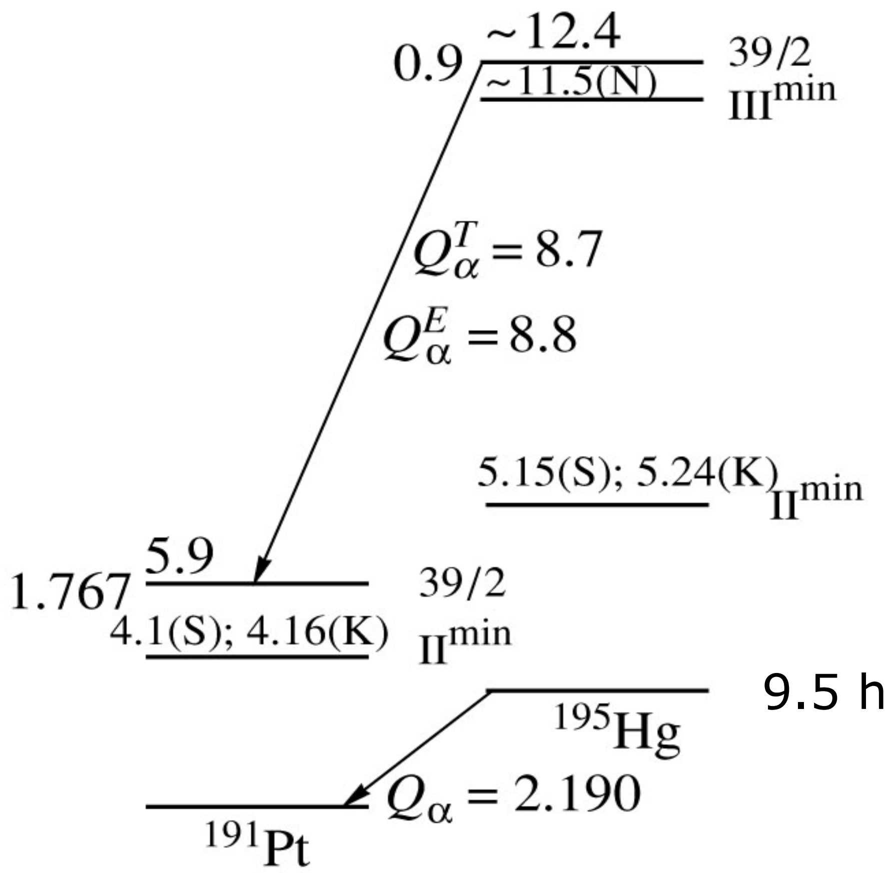
Eγ (keV)
250
225
200
175
150
125
100
75
50
25
0
0
10
20
30
40
50
60
J(J+1)-(J-1)J

$$E_x = 4.42xJ(J+1)$$

The 8.6 MeV α-particles decay very retardly to a high spin SD state. It cannot be a SD to a SD transition which decays very enhancely.

Int. J. Mod. Phys. E10 (2001) 185-208

Consistent interpretation: A transition from a high spin Hyperdeformed (HD) state to a high spin Superdeformed state.

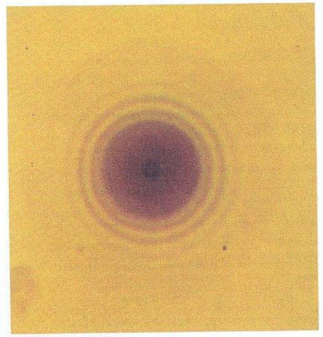


These observations motivated us to search for long-lived isomeric states in natural materials.

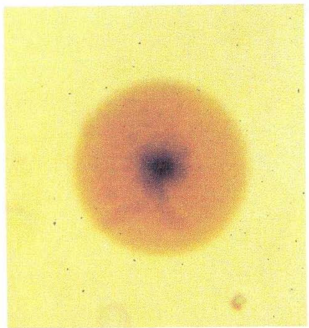
There are some radioactive decays seen in natural materials that were hitherto unexplained, but can be understood if one assume that long-lived isomeric states exist in nature.

^{238}U (4.2 MeV α) ^{234}Th (β) ^{234}Pa (β) ^{234}U (4.8 MeV α) ^{230}Th (4.7 MeV α) ^{226}Ra (4.8 MeV α) ^{222}Rn (5.5 MeV α) ^{218}Po (6.0 MeV α) ^{214}Pb (β) ^{214}Bi (β) ^{214}Po (7.7 MeV α) ^{210}Pb (β) ^{210}Bi (β) ^{210}Po (5.3 MeV α).

$t_{1/2}$ (^{238}U) = 4.5x10^9 y

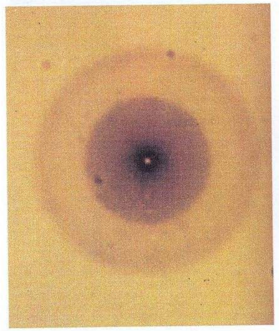


^{238}U



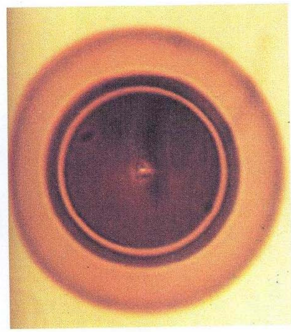
^{210}Po

$t_{1/2}$ (^{210}Po) = 138 d
$t_{1/2}$ (^{210}Pb) = 22 y (parent)



^{214}Po

$t_{1/2}$ (^{214}Po) = 164 μs
$t_{1/2}$ (^{214}Pb) = 26.8 m (parent)



^{218}Po

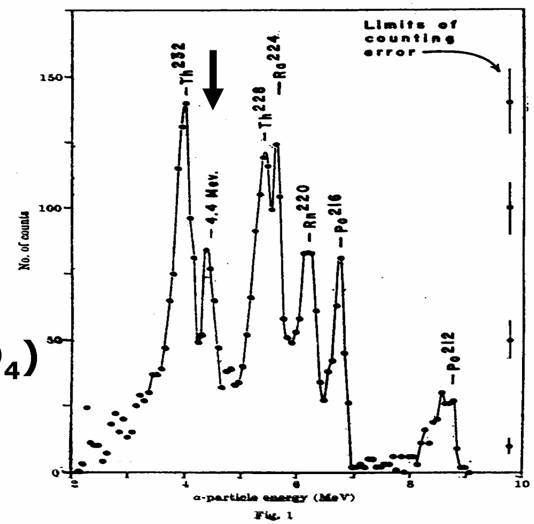
$t_{1/2}$ (^{218}Po) = 3 m

(Pictures from: R. V. Gentry, Creation's Tiny Mystery, Earth Science Associates Knoxville, Tennessee)

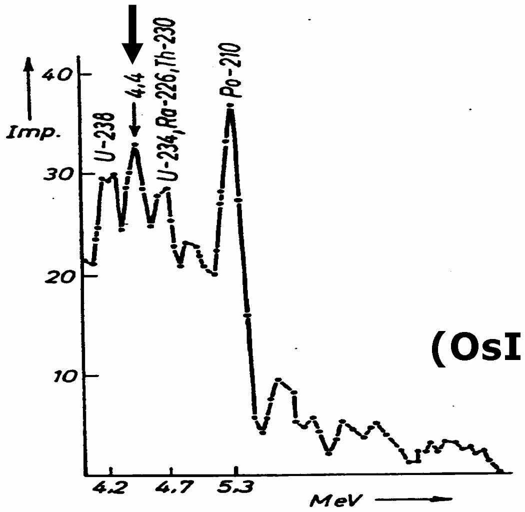
Long-lived isomeric states in the region around P o which decay by EC or beta particles can consistently interpret these puzzling halos.

The second phenomenon is the observation in various materials of a low energy α-particle group of 4.5 MeV

Thorite ($ThSiO_4$) from granite

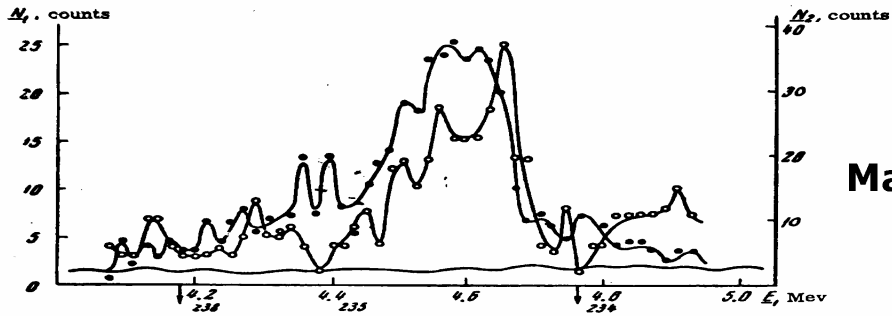


Cherry, Richardson & Adams
Nature **202**, 640 (1964)



Meier et al., Z. Naturforsch.
25 a, 79 (1970)

(OsIr)



Magnetite (Fe_3O_4) from Trans-caucacasian monazite

Cherdyntsev, Zverev, Kuptsov & Kislitsina
Geokhimiya, 395 (1968).

Distilled from strong nitric acid

Based on chemical properties it was assumed that the 4.5 MeV group is due to decay of an isotope of Hs (element 108, eka-Os)

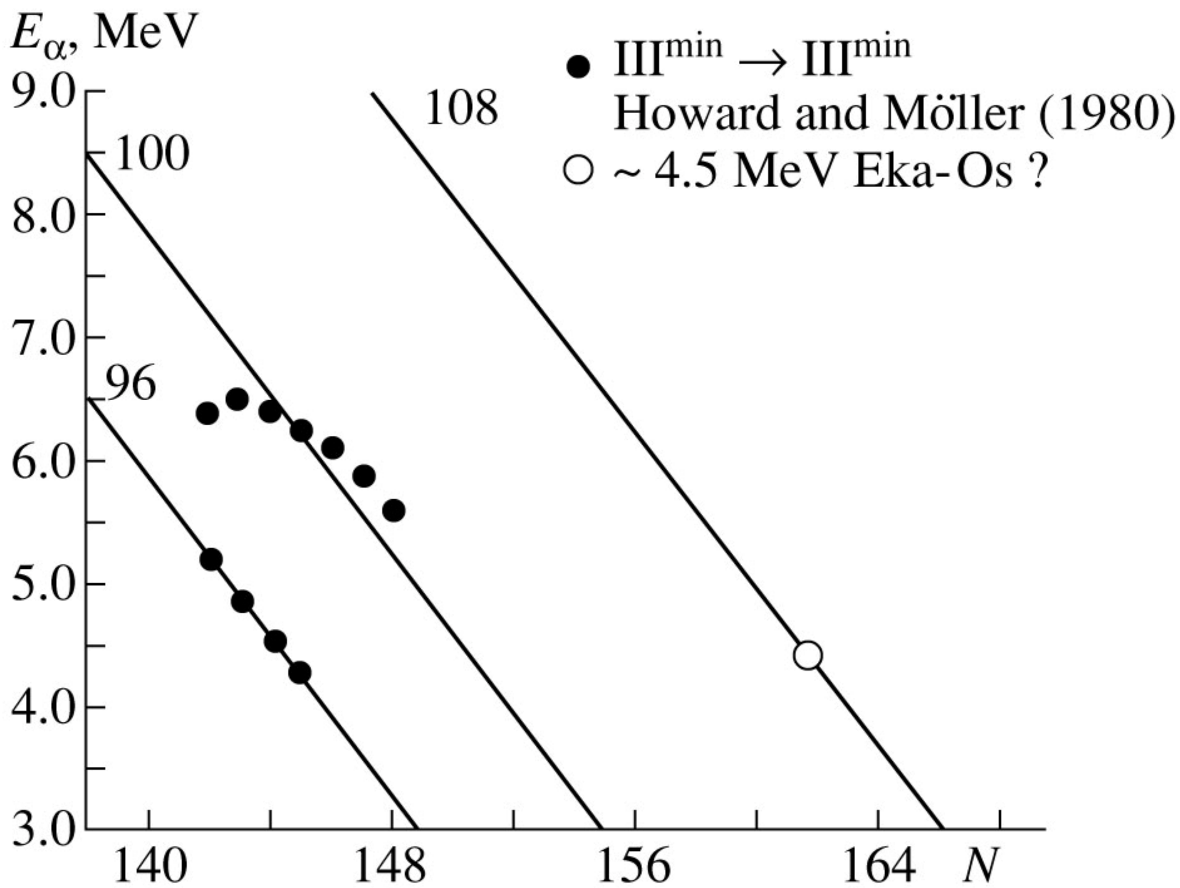
$t_{1/2} \approx 10^8$ y

However, the energy is too low (Normal energies are around 9 MeV, with $t_{1/2}$ ms to sec.) and it passes the barrier too fast

$t_{1/2}(\mathrm{Cal}) \approx 10^{16}$ y

Enhancement: 10^8

Consistent interpretation is obtained if one assumes HD to HD transition



Int. J. Mod. Phys. E12 (2003) 661

Table 5. Calculated half-lives for hyperdeformed to hyperdeformed α-particle transition of 4.5 MeV from ^{271}Hs assuming various deformation parameters [24]

β_2	β_3	β_4	$t_{1/2}$, yr
1.2[a]	0.0[b]	0.0[a]	1.8×10^{11}
1.2[a]	0.19[c]	0.0	4.6×10^{9}
0.85[d]	0.35[d]	0.18[d]	1.3×10^{8}

Most experiments searched for SHE in nature by looking for fission activities.

However: since $dN/dt = -(1/\tau)xN$

Then for $\tau \geq 10^8$ y one needs about 10^8 nuclei in order to see 1 dis/y.

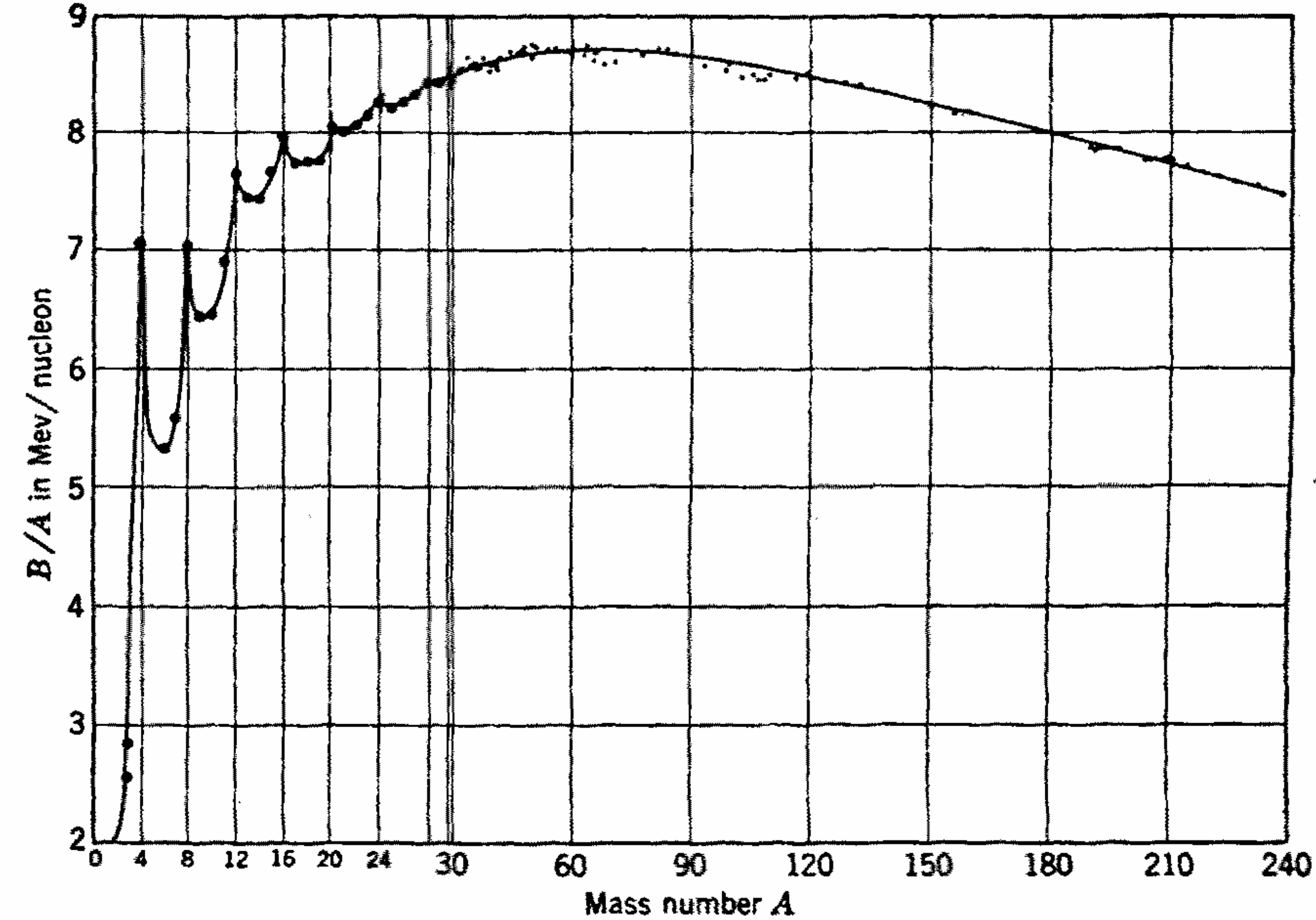
On the other hand for measuring N one needs about 10^4 - 10^5 atoms in an appropriate machine in order to see 1 event/s.

One way to measure the atomic mass number of nuclei is the AMS.

We tried it with negative results.

Another way is to measure the accurate mass of an atom by using a high resolution mass spectrometer that is able to separate between the mass of an atom and the masses of molecules of the same mass number.

$$M_A = ZxM_H + NxM_n - BE$$



The mass of any molecule (except for multi-hydrogen molecules or multi-Li, Be and B molecules) is lower than the mass of an atom with the same mass number.

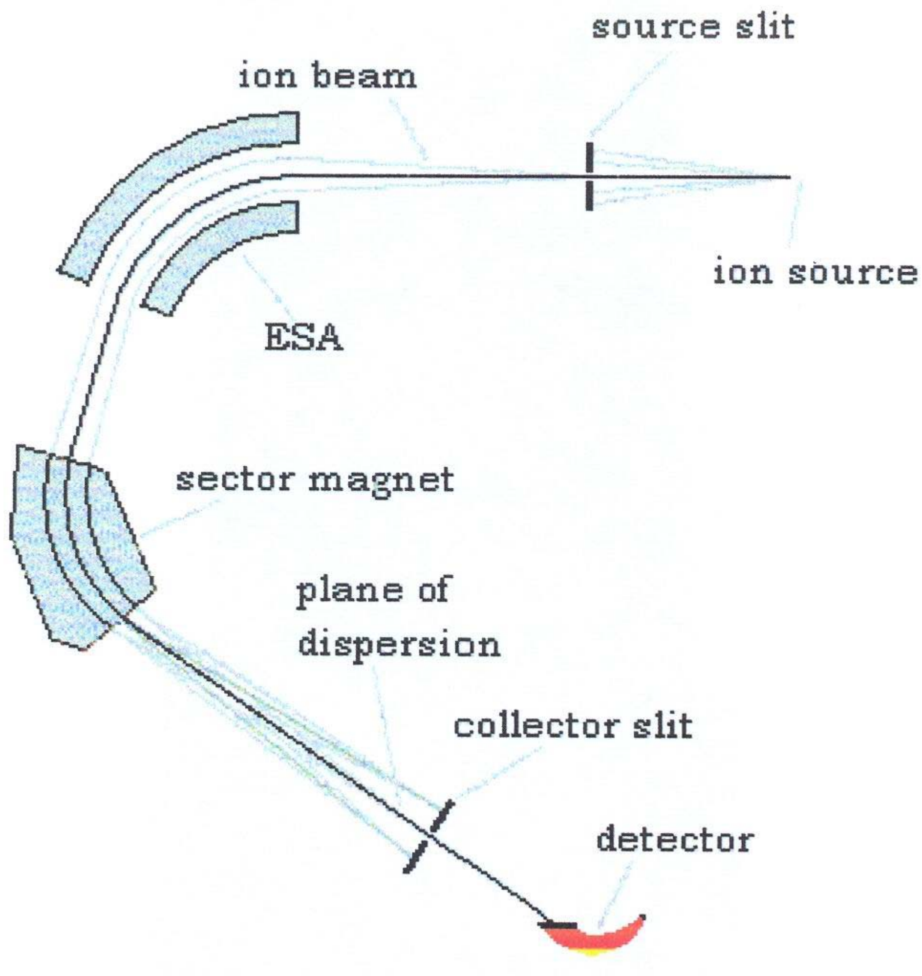
Inductively Coupled Plasma-Sector Field Mass Spectrometer (ICP-SFMS)

Plasma source at
6000-8000 K

Mostly atoms
from the source

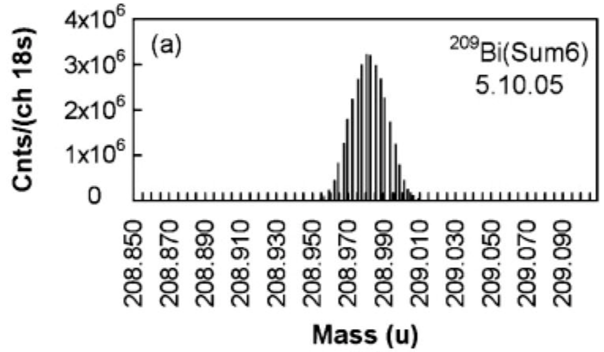
Studied material:
Solution

M/ΔM = 4000

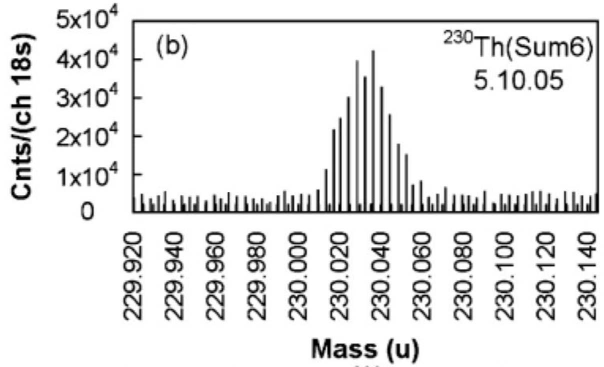


First: Measured neutron-deficient nuclei from pure Th solution

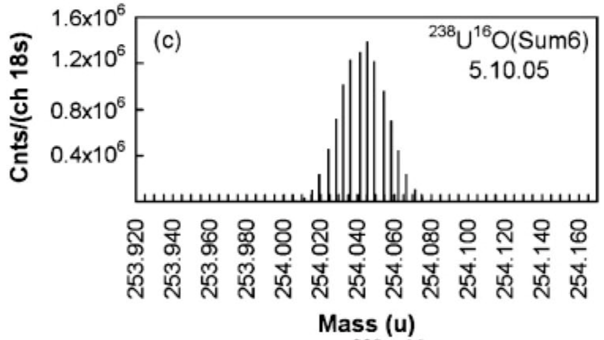
FWHM = 0.030 u



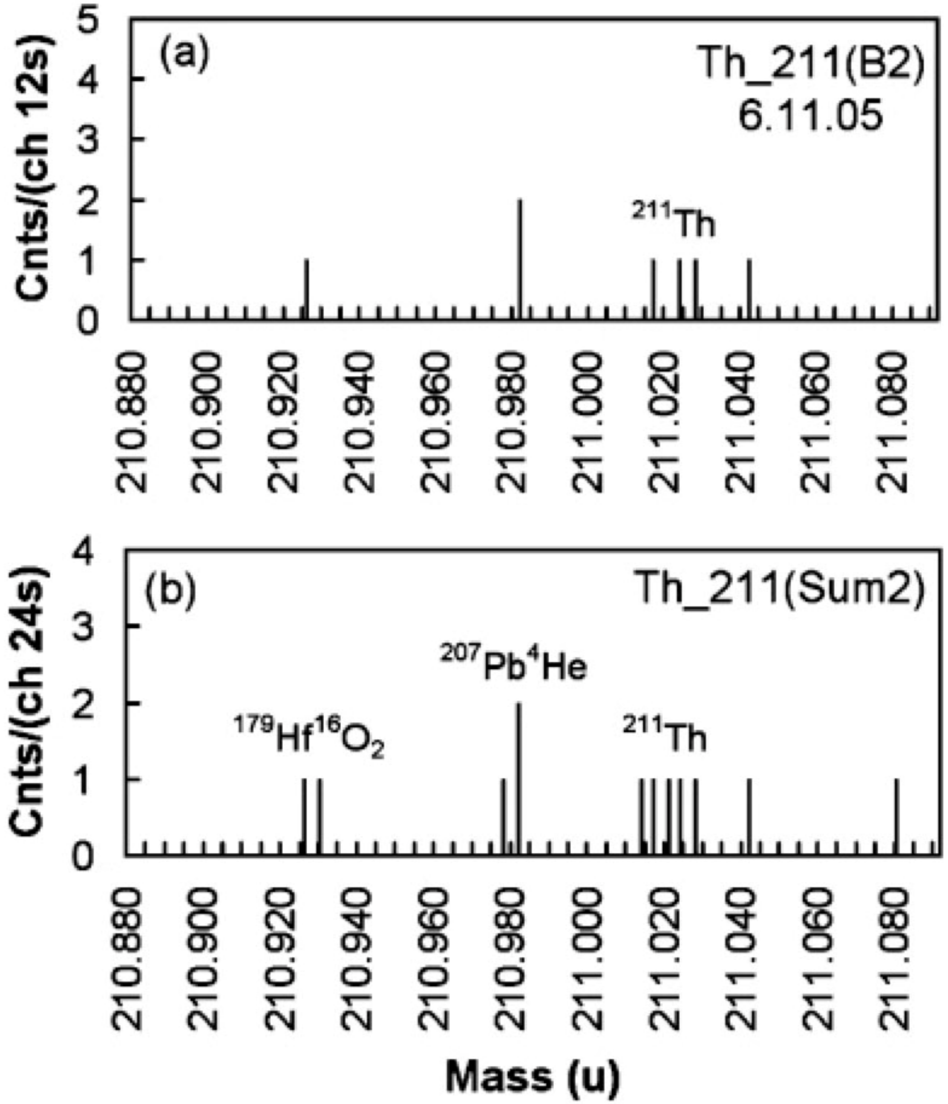
$M_{exp.}$(c.m.) = 208.981 u; M(^{209}Bi) = 208.980 u



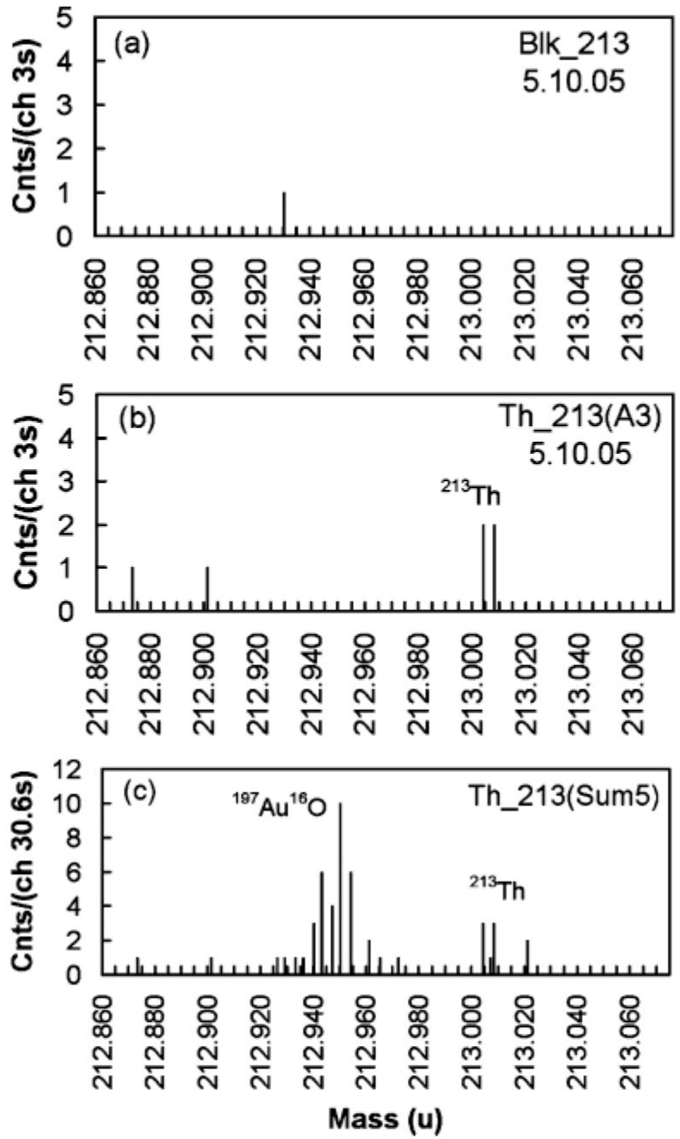
$M_{exp.}$(c.m.) = 230.035 u; M(^{230}Th) = 230.033 u



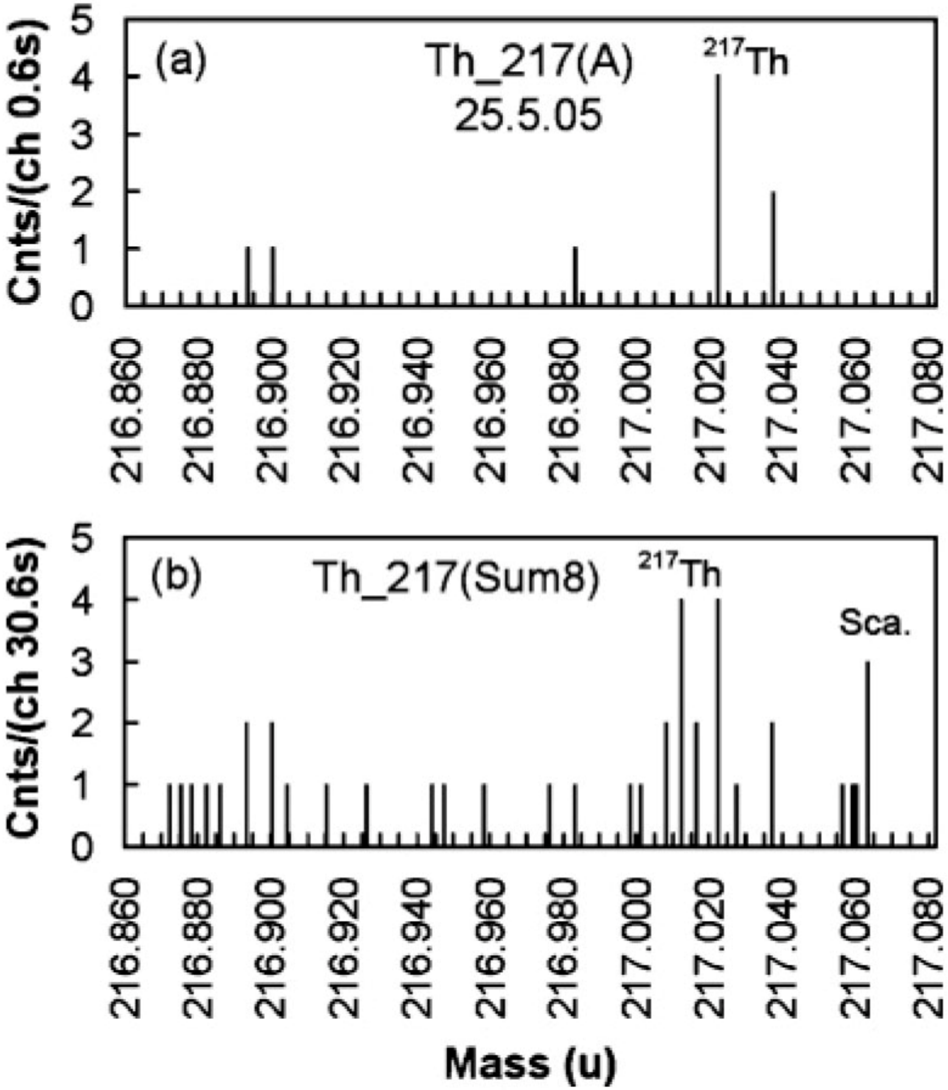
$M_{exp.}$(c.m.) = 254.043 u; M(^{238}U^{16}O) = 254.045 u

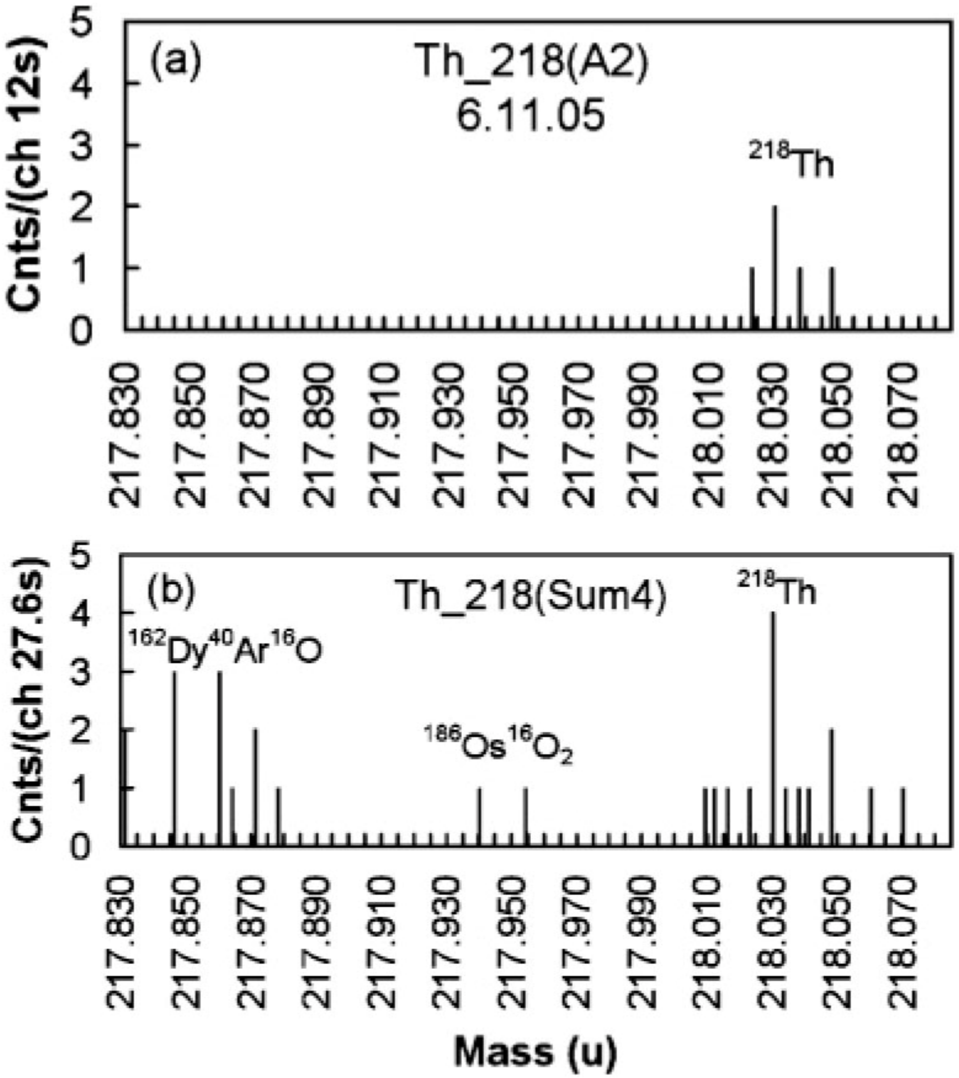


(a)
Th_211(B2)
6.11.05
211Th
Cnts/(ch 12s)
5
4
3
2
1
0
210.880
210.900
210.920
210.940
210.960
210.980
211.000
211.020
211.040
211.060
211.080
(b)
Th_211(Sum2)
207Pb4He
179Hf16O2
211Th
Cnts/(ch 24s)
4
3
2
1
0
210.880
210.900
210.920
210.940
210.960
210.980
211.000
211.020
211.040
211.060
211.080
Mass (u)



(a)
Blk_213
5.10.05
Cnts/(ch 3s)
(b)
Th_213(A3)
5.10.05
213Th
Cnts/(ch 3s)
(c)
197Au16O
Th_213(Sum5)
213Th
Cnts/(ch 30.6s)
212.860
212.880
212.900
212.920
212.940
212.960
212.980
213.000
213.020
213.040
213.060
Mass (u)


(a)
Th_217(A)
25.5.05
217Th
Cnts/(ch 0.6s)
(b)
Th_217(Sum8)
217Th
Sca.
Cnts/(ch 30.6s)
Mass (u)



(a)
Th_218(A2)
6.11.05
218Th
Cnts/(ch 12s)
(b)
Th_218(Sum4)
218Th
162Dy40Ar16O
186Os16O2
Cnts/(ch 27.6s)
Mass (u)

TABLE I. Summary of results of mass measurements and comparison with the known masses of the various Th isotopes.

Mass number	No. of events	No. of meas.	$P_{acc.}$	$M_{exp.}$[a] (average)	$M_{g.s.}$ of Th isotope[b]
211	5	2	5×10^{-4}	211.021	211.015
213	9	5	6×10^{-7}	213.012	213.013
217	15	8	9×10^{-7}	217.018	217.013
218	13	4	6×10^{-6}	218.021	218.013

[a]The uncertainties in mass are estimated to be ±0.015 u.

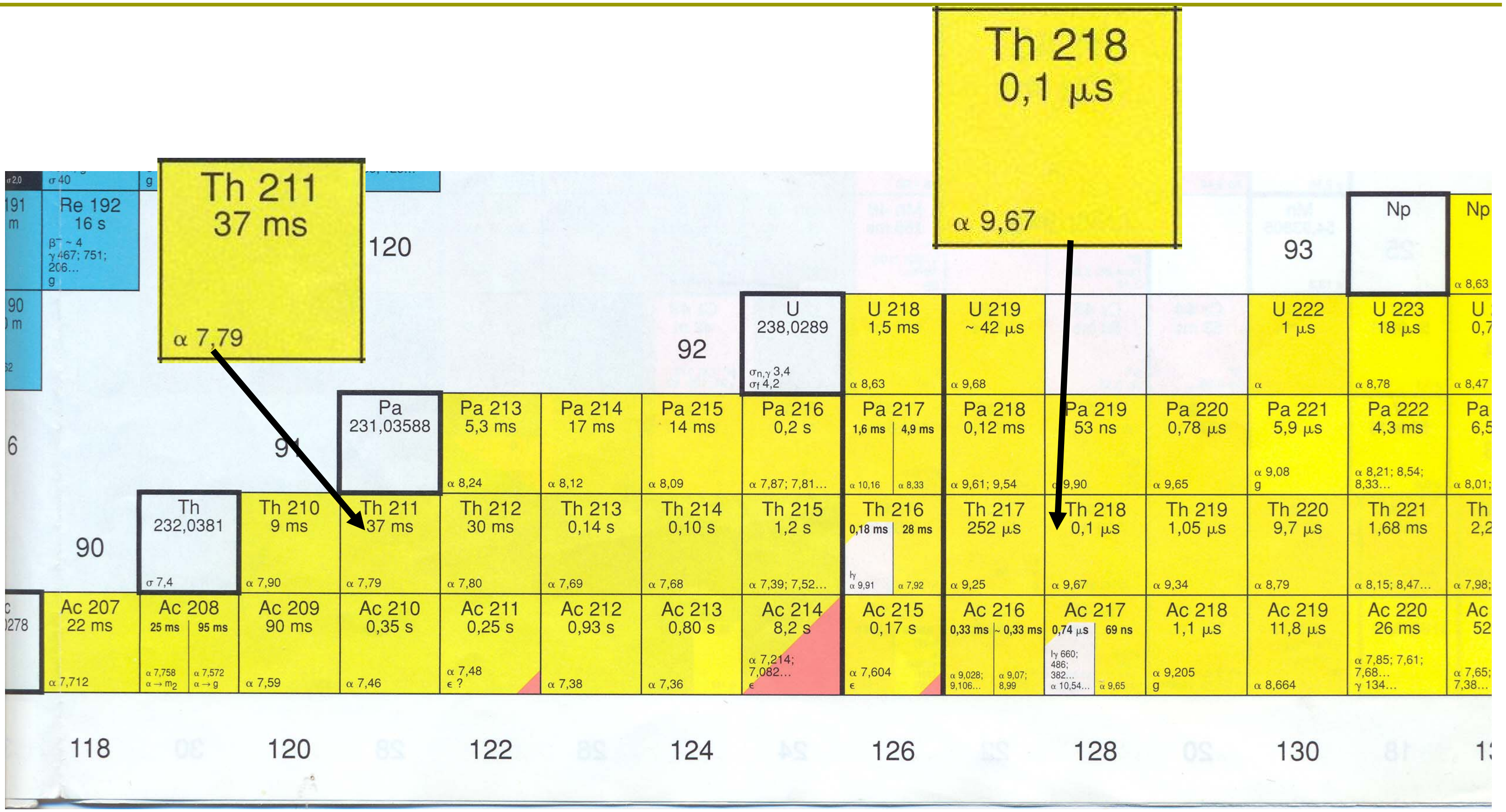
[b]Reference [21].

All together we saw 42 events in 19 independent measurements.

The relative abundance of these isotopes compared to ^{232}Th is (1 -10)x10^{-11}

(2-20)x10^{-16} of the solution.

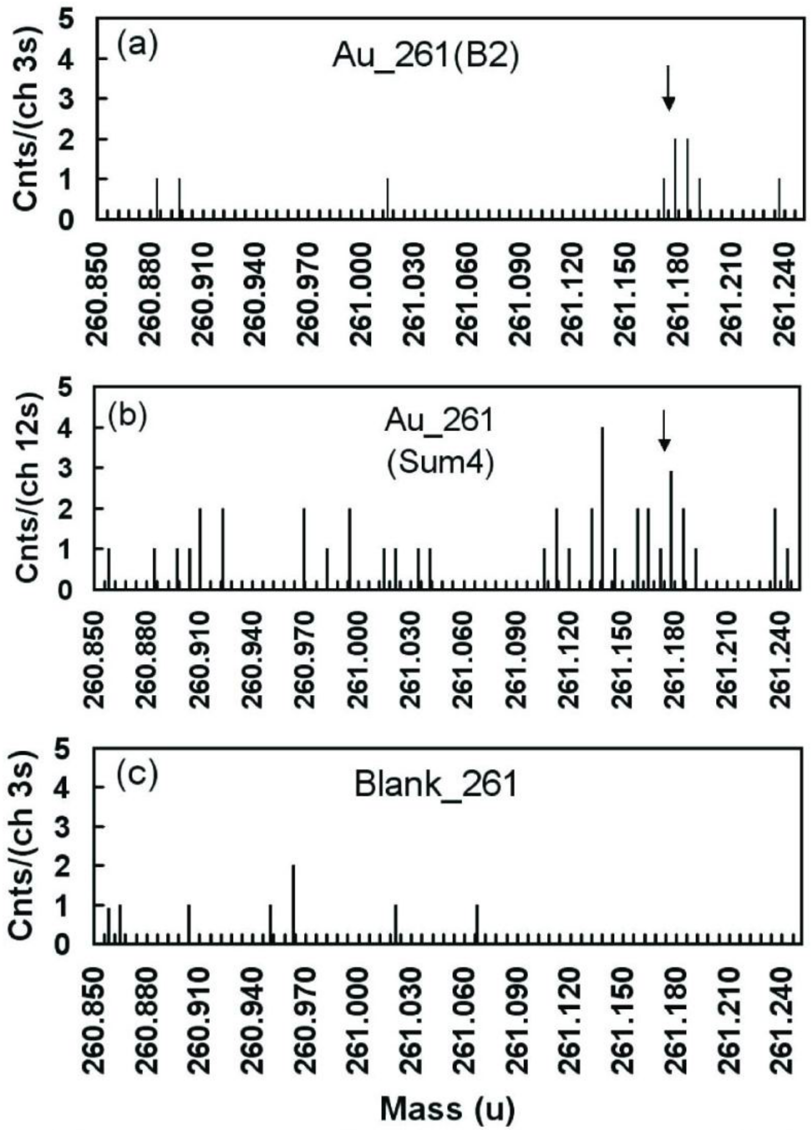
If the terrestrial concentration of these isotopes were initially the same as of ^{232}Th then $t_{1/2} \geq 10^8$ y.

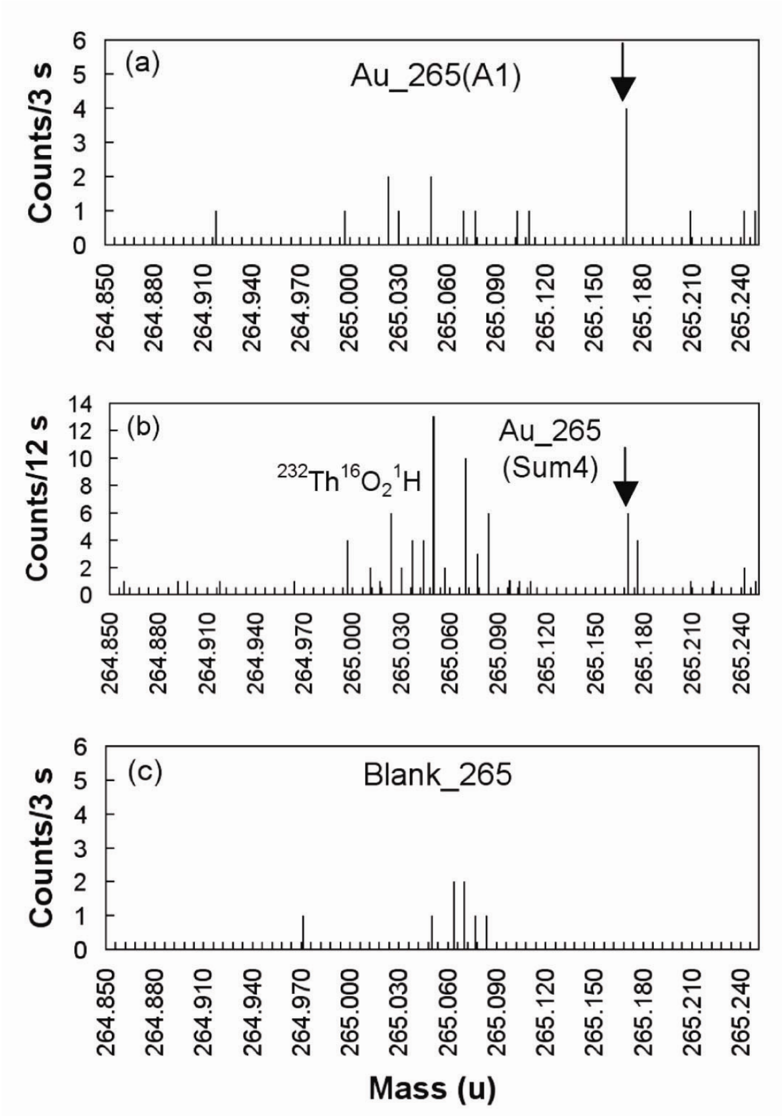

Th 211
37 ms
α 7,79
Th 218
0,1 µs
α 9,67

Conclusion: Long-lived isomeric states with half-lives 10^{16} to 10^{22} longer than their corresponding g.s. have been found in the neutron-deficient $^{211,213,217,218}Th$ nuclei.

PRC 76, 021303(R) (2007)

Our second experiment was to search for long-lived isomeric states in pure Au solution looking for high masses, assuming that if Rg (eka-Au, element 111) exists in nature it may be found together with Au.


(a)
Au_261(B2)
Cnts/(ch 3s)
(b)
Au_261
(Sum4)
Cnts/(ch 12s)
(c)
Blank_261
Cnts/(ch 3s)
0
1
2
3
4
5
260.850
260.880
260.910
260.940
260.970
261.000
261.030
261.060
261.090
261.120
261.150
261.180
261.210
261.240
Mass (u)



(a)
Au_265(A1)
Counts/3 s
(b)
Au_265
(Sum4)
$^{232}Th^{16}O_2{}^1H$
Counts/12 s
(c)
Blank_265
Counts/3 s
Mass (u)

TABLE I: Summary of results of mass measurements and comparison with the predicted masses of ^{261}Rg and ^{265}Rg.

Mass no.	Fig. no.	No. of events	$P_{acc.}$	$M_{c.m.}^{exp.\ a}$	Mass of Rg isotope[b]
261	2(a)	6	$5x10^{-7}$		
261	2(b)	22(18)	$3x10^{-6\ c}$	261.134[d]	261.154
265	3(a)	4	$3x10^{-7}$		
265	3(b)	10	$1x10^{-9}$	265.154	265.151

[a]The uncertainty in mass is estimated to be ±0.025 u.
[b]Average of predicted values, Refs. [3, 4, 5].
[c]Because of the different widths of the lines, the same value is obtained for 22 and 18 events lines.
[d]For 18 counts $M_{c.m.}^{exp.}$=261.142

All together we saw about 40 events in eight independent measurements

The relative abundance of these isotopes compared to ^{197}Au is (1 -10)x10^{-10} (2-20)x10^{-15} of the solution.

The chemical properties of Sg(106), Bh(107), Hs(108) and element 112

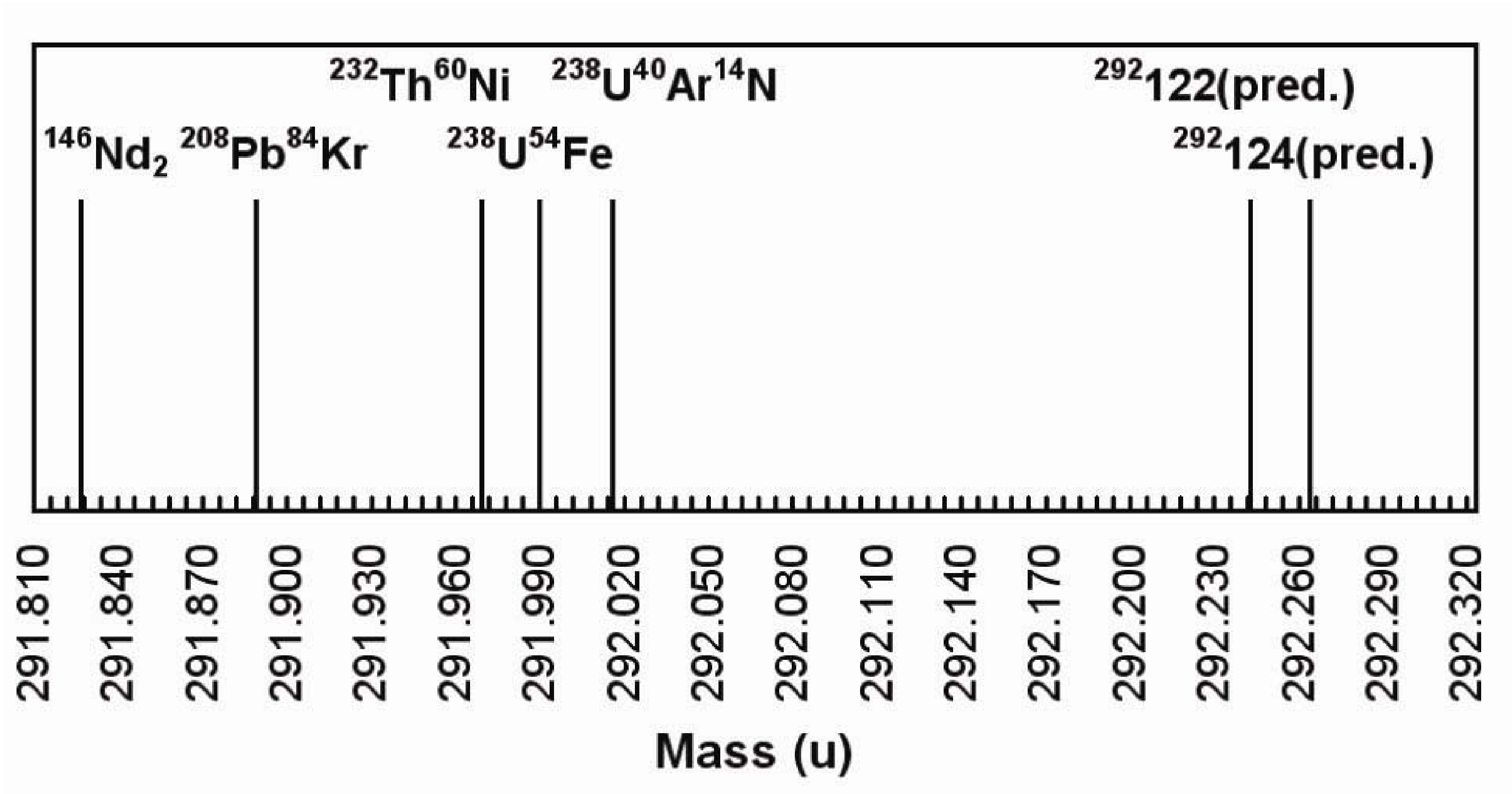
were found to be similar to those of their lighter homologues,

W, Re, Os, and Hg.

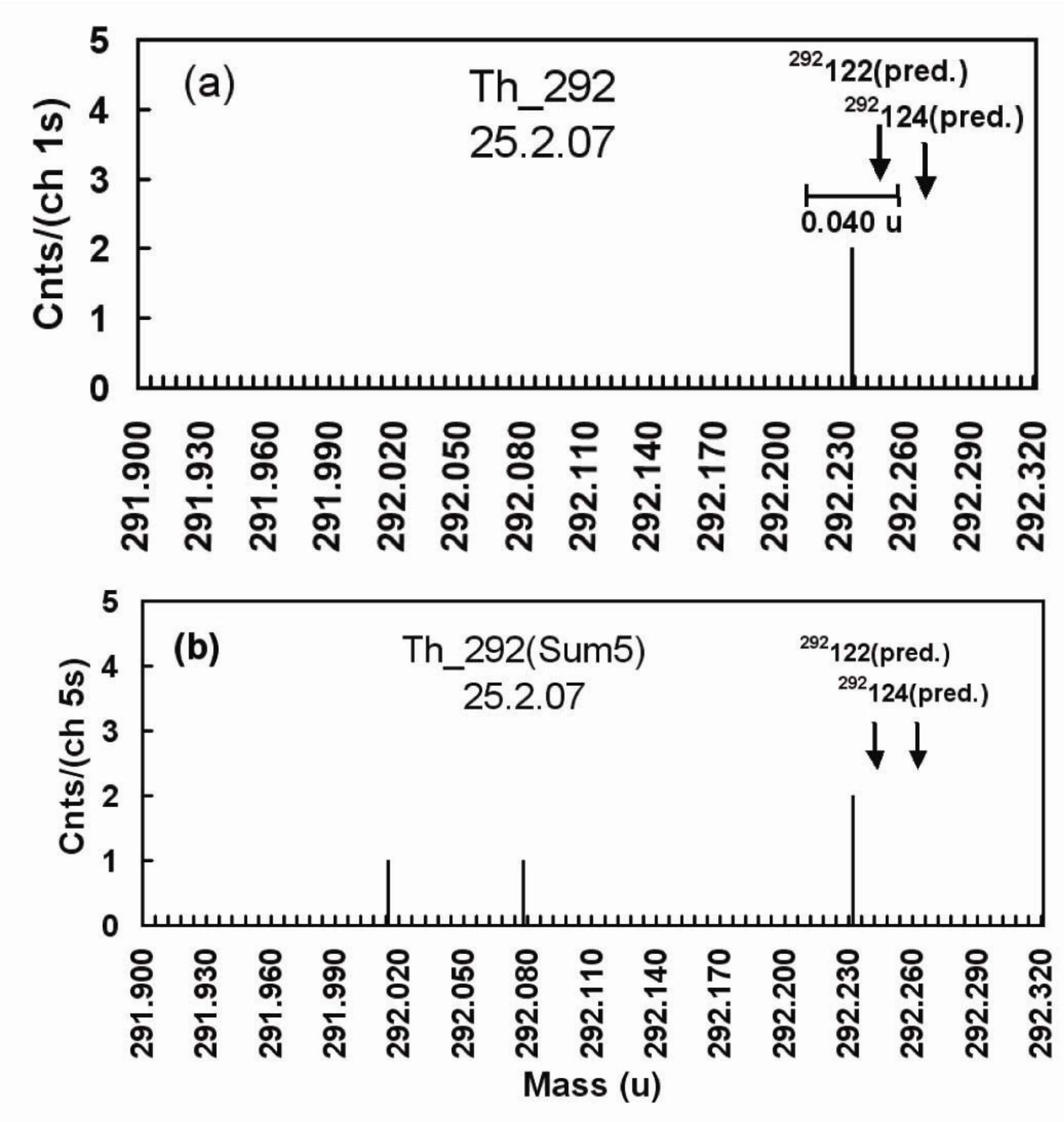
Therefore one may assume that the observed A=261 and 265 nuclei are ^{261}Rg and ^{265}Rg (element 111).

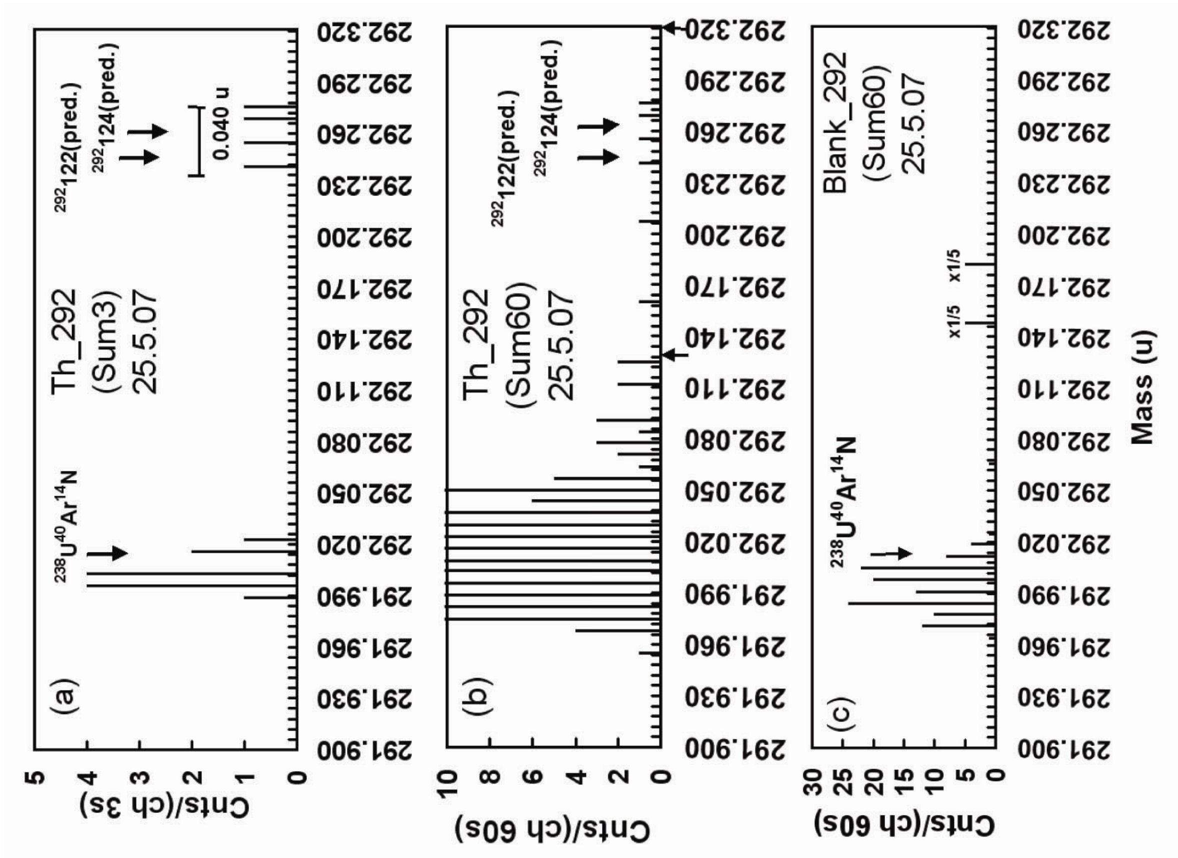
Third experiment:

Search for SHE in Th solution but at high masses from 287 to 294, looking for suuperactinide nuclei. According to the extended periodic table of Seaborg, elements 122 and 124 are placed as eka-Th and eka-U, respectively.

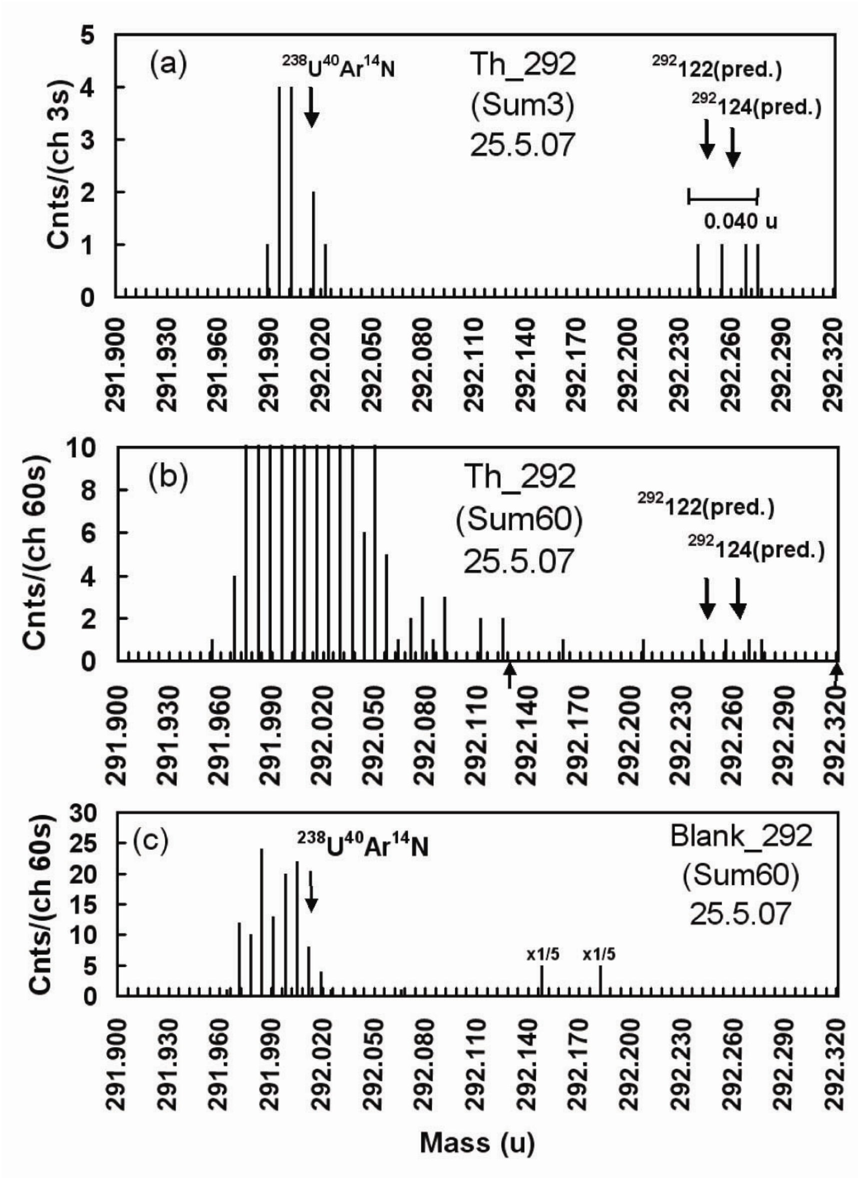


232Th60Ni
238U40Ar14N
292122(pred.)
146Nd2
208Pb84Kr
238U54Fe
292124(pred.)
291.810
291.840
291.870
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
Mass (u)

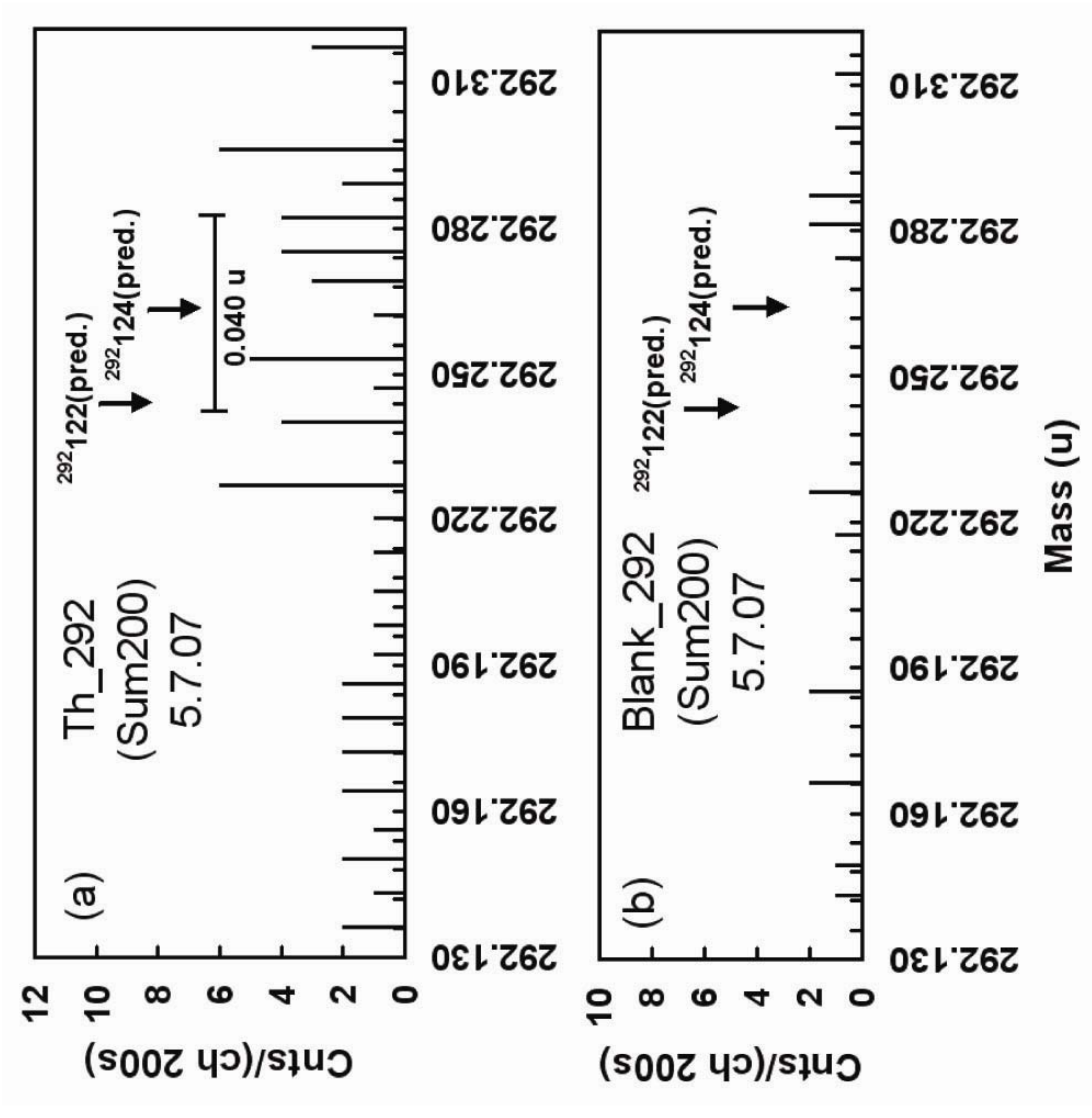

(a)
Th_292
25.2.07
292122(pred.)
292124(pred.)
0.040 u
Cnts/(ch 1s)
(b)
Th_292(Sum5)
25.2.07
292122(pred.)
292124(pred.)
Cnts/(ch 5s)
Mass (u)



(a)
Th_292
(Sum3)
25.5.07
238U40Ar14N
292122(pred.)
292124(pred.)
0.040 u
Cnts/(ch 3s)
(b)
Th_292
(Sum60)
25.5.07
292122(pred.)
292124(pred.)
Cnts/(ch 60s)
(c)
Blank_292
(Sum60)
25.5.07
238U40Ar14N
x1/5
x1/5
Cnts/(ch 60s)
Mass (u)



In the next experiment we focus on the limited region of 292.13u u to 292.32u



(a)
Th_292
(Sum200)
5.7.07
292122(pred.)
292124(pred.)
0.040 u
Cnts/(ch 200s)
(b)
Blank_292
(Sum200)
5.7.07
292122(pred.)
292124(pred.)
Cnts/(ch 200s)
Mass (u)

$M_{exp.} = 292.262 \pm 0.030$ u

Predictions (KTUY05; LMZ01)

For $^{292}121$ to $^{292}126$ are

292.236 u to 292.291 u

Abundance (relative to ^{232}Th):

$(1\text{-}10)\times10^{-12}$

$t_{1/2} \geq 10^8$ y

Chemical arguments:

a) We used pure Th solution.

b) The atomic configuration of Th is $6d_{3/2}^{2}7s^{2}$ and its separation is based on its stable 4^{+} oxidation state.

c) The accurate predicted (U. Kaldor) atomic configuration of eka-Th (Z = 122) is $8s^{2}7d_{3/2}8p_{1/2}$. It is also expected to form a stable 4^{+} state.

d) Element 121 has only three electrons outside the closed shells of element 118 (eka-Rn) and it is not likely that it will form a stable 4^+ state,

e) Elements above Z=122 have more electrons. They can form 4^+ oxidation state, but also higher oxidation states.

f) If element 122 exists in nature together with Th, it is reasonable to assume that it followed Th in the chemical separation, and showed up in our measurements. However the possibility that A =292 Nucleus belongs to an element of somewhat higher Z cannot be excluded.

The predicted half-lives of nuclei around ^{292}122 are:

$t_{1/2}$ (pred.) = 10^{-6} - 10^{-8} s.

$t_{1/2}$ (exp.) $\geq 10^{8}$ y

Conclusion: What we found is an isomeric state in the nucleus

A = 292 and Z $\cong$ 122

Pred.: Möller, Nix, Kratz, ADNDT (1997)

122/292

≥ 10⁸ y

112/272

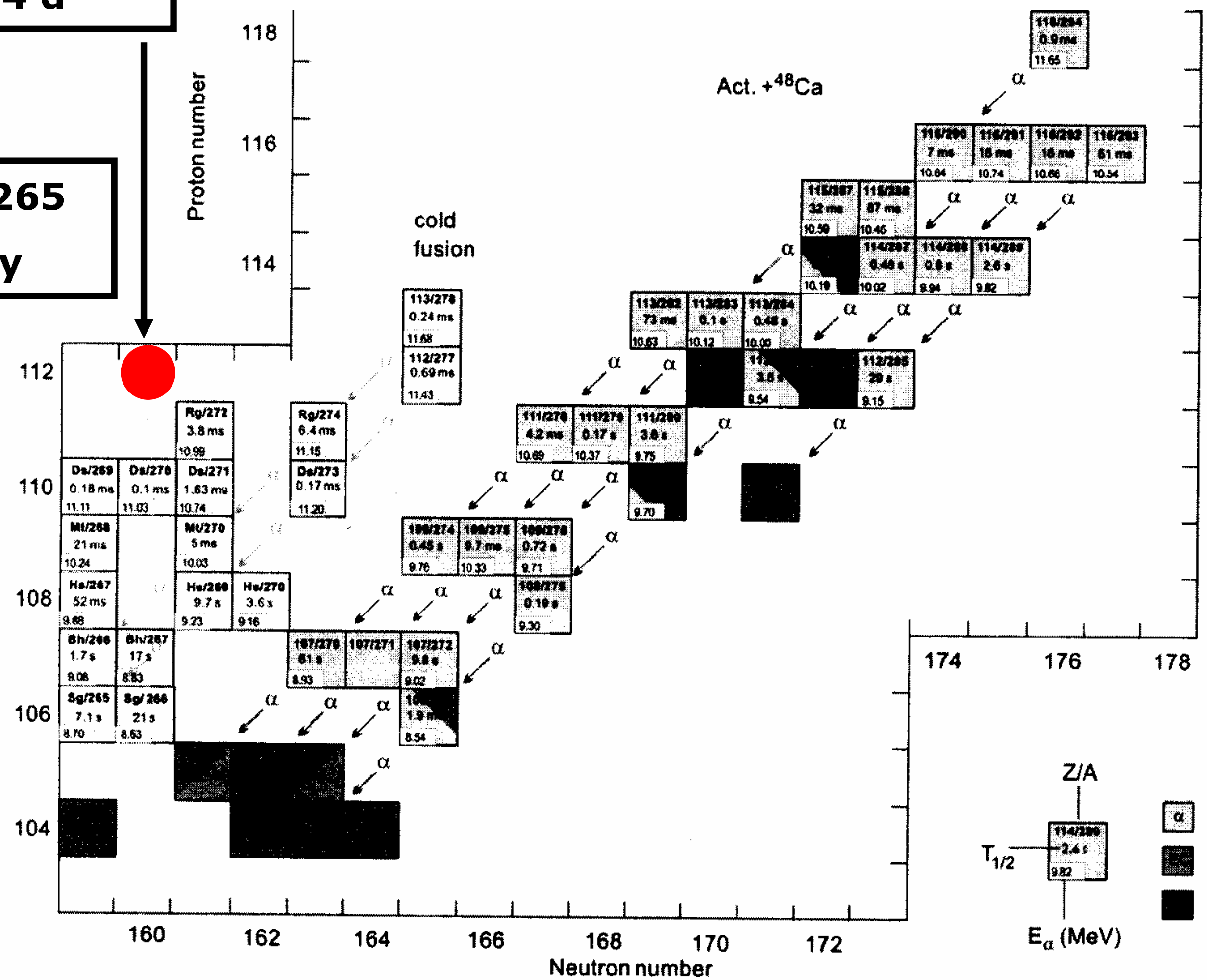
~14 d

111/261

≥ 10⁸ y

111/265

≥ 10⁸ y



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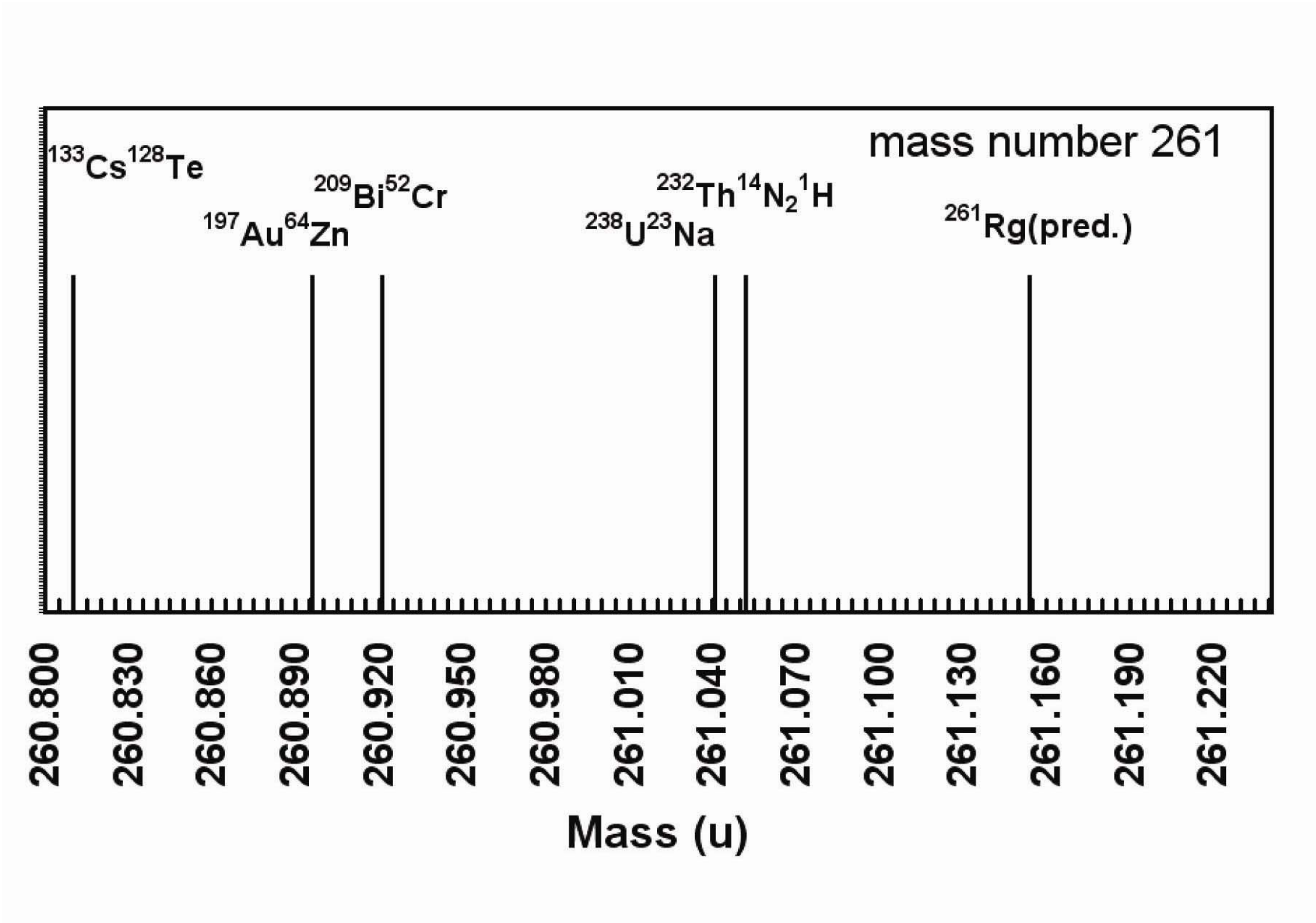
Thank You for Your Attention



Predictions: a) Möller *et al. (1995)* b) Koura *et al. (2005)*

c) Liran, Marinov and Zeldes (2000)